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PATENT TRADEMARK OFFICE

CHAPTER II

TRANSMITTAL LETTER
TO THE UNITED STATES ELECTED OFFICE (EO/US)
(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/IB00/00290	17 MARCH 2000	18 MARCH 1999

TITLE OF INVENTION

METHOD OF OBTAINING METHYL ALCOHOL FROM RAW GAS AND INSTALLATION FOR CARRYING OUT SAID METHOD

APPLICANT(S)

Emil A. WIESER-LINHART

Box PCT**Assistant Commissioner for Patents****Washington D.C. 20231****ATTENTION: EO/US****CERTIFICATION UNDER 37 C.F.R. 1.10****(Express Mail label number is mandatory.)**(Express Mail certification is optional.)*

I hereby certify that this correspondence and the documents referred to as attached therein are being deposited with the United States Postal Service on this date SEPTEMBER 4, 2001, in an envelope as "Express Mail Post Office to Addressee," Mailing Label Number EL728214985US, addressed to the: Assistant Commissioner for Patents, Washington, D.C. 20231.

JENNIFER RASHKIN*(type or print name of person mailing paper)**Signature of person mailing paper*

WARNING: Certificate of mailing (first class) or facsimile transmission procedures of 37 C.F.R. 1.8 cannot be used to obtain a date of mailing or transmission for this correspondence.

***WARNING:** Each paper or fee filed by "Express Mail" **must** have the number of the "Express Mail" mailing label placed thereon prior to mailing, 37 C.F.R. 1.10(b).
 "Since the filing of correspondence under § 1.10 without the Express Mail mailing label thereon is an oversight that can be avoided by the exercise of reasonable care, requests for waiver of this requirement will **not** be granted on petition." Notice of Oct. 24, 1996, 60 Fed. Reg. 56,439, at 56,442.

NOTE: *The completion of those filing requirements that can be made at a time later than 30 months from the priority date results from the Commissioner exercising his judgment under the authority granted under 35 USC 371(d). The filing receipt will show the actual date of receipt of the last item completing the entry into the national phase. See 37 C.F.R. §1.491 which states: "An international application enters the national state when the applicant has filed the documents and fees required by 35 USC 371(c) within the periods set forth in § 1.494 and § 1.495."*

WARNING: *Where the items are those which can be submitted to complete the entry of the international application into the national phase are subsequent to 30 months from the priority date the application is still considered to be in the international state and if mailing procedures are utilized to obtain a date the express mail procedure of 37 C.F.R. §1.10 must be used (since international application papers are not covered by an ordinary certificate of mailing - See 37 C.F.R. §1.8.*

NOTE: *Documents and fees must be clearly identified as a submission to enter the national state under 35 USC 371 otherwise the submission will be considered as being made under 35 USC 111. 37 C.F.R. § 1.494(f).*

1. Applicant herewith submits to the United States Elected Office (EO/US) the following items under 35 U.S.C. 371:
 - a. ☒ [X] This express request to immediately begin national examination procedures (35 U.S.C. 371(f)).
 - b. ☒ [X] The U.S. National Fee (35 U.S.C. 371(c)(1)) and other fees (37 C.F.R. § 1.492) as indicated below:

2. Fees

CLAIMS FEE	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULA- TIONS
[]*	TOTAL CLAIMS	13- 20 =		x \$ 18.00 =	\$
	INDEPENDENT CLAIMS	1- 3 =		x \$ 80.00 =	
	MULTIPLE DEPENDENT CLAIM(S) (if applicable) + \$270.00				
BASIC FEE**	<input type="checkbox"/> U.S. PTO WAS INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where an international preliminary examination fee as set forth in § 1.482 has been paid on the international application to the U.S. PTO: <input type="checkbox"/> and the international preliminary examination report states that the criteria of novelty, inventive step (non-obviousness) and industrial activity, as defined in PCT Article 33(2) to (4) have been satisfied for all the claims presented in the application entering the national stage (37 CFR 1.492(a)(4)) \$100.00 <input type="checkbox"/> and the above requirements are not met (37 CFR 1.492(a)(1)) \$690.00 <input checked="" type="checkbox"/> U.S. PTO WAS NOT INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where no international preliminary examination fee as set forth in § 1.482 has been paid to the U.S. PTO, and payment of an international search fee as set forth in § 1.445(a)(2) to the U.S. PTO: <input type="checkbox"/> has been paid (37 CFR 1.492(a)(2)) \$710.00 <input type="checkbox"/> has not been paid (37 CFR 1.492(a)(3)) \$1,000.00 <input checked="" type="checkbox"/> where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 CFR 1.492(a)(5)) \$860.00				
	Total of above Calculations				= \$60.00
SMALL ENTITY	Reduction by ½ for filing by small entity, if applicable. Statement may also be filed. (note 37 CFR 1.9, 1.27, 1.28)				-
	Subtotal				\$60.00
	Total National Fee				\$860.00
	Fee for recording the enclosed assignment document \$40.00 (37 CFR 1.21(h)). (See Item 13 below). See attached "ASSIGNMENT COVER SHEET".				
TOTAL	Total Fees enclosed				\$860.00

*See attached Preliminary Amendment Reducing the Number of Claims.

- i. ☒ [X] A check in the amount of \$860.00 to cover the above fees is enclosed.
 ii. ☐ [] Please charge Account No. _____ in the amount of \$ _____.
 A duplicate copy of this sheet is enclosed.

****WARNING:** "To avoid abandonment of the application the applicant shall furnish to the United States Patent and Trademark Office not later than the expiration of 30 months from the priority date: * * * (2) the basic national fee (see § 1.492(a)). The 30-month time limit may not be extended." 37 C.F.R. § 1.495(b).

WARNING: If the translation of the international application and/or the oath or declaration have not been submitted by the applicant within thirty (30) months from the priority date, such requirements may be met within a time period set by the Office. 37 C.F.R. § 1.495(b)(2). The payment of the surcharge set forth in § 1.492(e) is required as a condition for accepting the oath or declaration later than thirty (30) months after the priority date. The payment of the processing fee set forth in § 1.492(f) is required for acceptance of an English translation later than thirty (30) months after the priority date. Failure to comply with these requirements will result in abandonment of the application. The provisions of § 1.136 apply to the period which is set. Notice of Jan. 3, 1993, 1147 O.G. 29 to 40.

3. ☒ [X] A copy of the International application as filed (35 U.S.C. 371(c)(2)):

NOTE: Section 1.495 (b) was amended to require that the basic national fee and a copy of the international application must be filed with the Office by 30 months from the priority date to avoid abandonment "The International Bureau normally provides the copy of the international application to the Office in accordance with PCT Article 20. At the same time, the International Bureau notifies applicant of the communication to the Office. In accordance with PCT Rule 47.1, that notice shall be accepted by all designated offices as conclusive evidence that the communication has duly taken place. Thus, if the applicant desires to enter the national stage, the applicant normally need only check to be sure the notice from the International Bureau has been received and then pay the basic national fee by 30 months from the priority date." Notice of Jan. 7, 1993, 1147 O.G. 29 to 40, at 35-36. See item 14c below.

- a. ☐ [] is transmitted herewith.
 b. ☐ [] is not required, as the application was filed with the United States Receiving Office.
 c. ☒ [X] has been transmitted
 i. ☒ [X] by the International Bureau.
 Date of mailing of the application (from form PCT/IB/308): _____.
 ii. ☐ [] by applicant on _____
 Date

4. ☒ [X] A translation of the International application into the English language (35 U.S.C. 371(c)(2)):
 a. ☒ [X] is transmitted herewith.
 b. ☐ [] is not required as the application was filed in English.
 c. ☐ [] was previously transmitted by applicant on _____
 Date
 d. ☐ [] will follow.

5. ☐ Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. 371(c)(3)):

NOTE: The Notice of January 7, 1993 points out that 37 C.F.R. § 1.495(a) was amended to clarify the existing and continuing practice that PCT Article 19 amendments must be submitted by 30 months from the priority date and this deadline may not be extended. The Notice further advises that: "The failure to do so will not result in loss of the subject matter of the PCT Article 19 amendments. Applicant may submit that subject matter in a preliminary amendment filed under section 1.121. In many cases, filing an amendment under section 1.121 is preferable since grammatical or idiomatic errors may be corrected." 1147 O.G. 29-40, at 36.

- a. ☐ are transmitted herewith.
b. ☐ have been transmitted
i. ☐ by the International Bureau.
Date of mailing of the amendment (from form PCT/IB/308): _____.
ii. ☐ by applicant on _____.
Date
c. ☒ have not been transmitted as
i. ☒ applicant chose not to make amendments under PCT Article 19.
Date of mailing of Search Report (from form PCT/ISA/210): 14 JUNE 2000.
ii. ☐ the time limit for the submission of amendments has not yet expired.
The amendments or a statement that amendments have not been made will be transmitted before the expiration of the time limit under PCT Rule 46.1.

6. ☒ A translation of the amendments to the claims under PCT Article 19 (38 U.S.C. 371(c)(3)):

- a. ☐ is transmitted herewith.
b. ☐ is not required as the amendments were made in the English language.
c. ☒ has not been transmitted for reasons indicated at point 5(c) above.

7. ☒ A copy of the international examination report (PCT/IPEA/409)

- ☒ is transmitted herewith.
☐ is not required as the application was filed with the United States Receiving Office.

8. ☐ Annex(es) to the international preliminary examination report

- a. ☐ is/are transmitted herewith.
b. ☐ is/are not required as the application was filed with the United States Receiving Office.

9. ☐ A translation of the annexes to the international preliminary examination report

- a. ☐ is transmitted herewith.
b. ☐ is not required as the annexes are in the English language.

10. ☒ An oath or declaration of the inventor (35 U.S.C. 371(c)(4)) complying with 35 U.S.C. 115
- a. ☐ was previously submitted by applicant on _____
Date
- b. ☐ is submitted herewith, and such oath or declaration
- i. ☐ is attached to the application.
- ii. ☐ identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or 3(c) and 5(b); and states that they were reviewed by the inventor as required by 37 C.F.R. 1.70.
- c. ☒ will follow.

Other document(s) or information included:

11. ☒ An International Search Report (PCT/ISA/210) or Declaration under PCT Article 17(2)(a):
- a. ☒ is transmitted herewith.
- b. ☐ has been transmitted by the International Bureau.
Date of mailing (from form PCT/IB/308): _____
- c. ☐ is not required, as the application was searched by the United States International Searching Authority.
- d. ☐ will be transmitted promptly upon request.
- e. ☐ has been submitted by applicant on _____
Date
12. ☒ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98:
- a. ☐ is transmitted herewith.
Also transmitted herewith is/are:
☐ Form PTO-1449 (PTO/SB/08A and 08B).
☐ Copies of citations listed.
- b. ☒ will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. 371(c).
- c. ☐ was previously submitted by applicant on _____
Date
13. ☐ An assignment document is transmitted herewith for recording.

A separate ☐ "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or ☐ FORM PTO 1595 is also attached.

14. ☒ Additional documents:
- a. ☒ Copy of request (PCT/RO/101)
 - b. ☒ International Publication No. WO 00/55347
 - i. ☒ Specification, claims and drawing
 - ii. ☐ Front page only
 - c. ☐ Preliminary amendment (37 C.F.R. § 1.121)
 - d. ☒ Other

FORM PCT/IPEA/401; FORM PCT/IPEA/416; FORM PCT/IB/301

15. ☒ The above checked items are being transmitted
- a. ☒ before 30 months from any claimed priority date.
 - b. ☐ after 30 months.
16. ☐ Certain requirements under 35 U.S.C. 371 were previously submitted by the applicant on _____, namely:

AUTHORIZATION TO CHARGE ADDITIONAL FEES

WARNING: *Accurately count claims, especially multiple dependent claims, to avoid unexpected high charges if extra claims are authorized.*

NOTE: *"A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time. An authorization to charge all required fees, fees under § 1.17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission. Submission of the fee set forth in § 1.17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 C.F.R. § 1.136(a)(3).*

NOTE: *"Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts; amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 C.F.R. § 1.26(a).*

- ☒ The Commissioner is hereby authorized to charge the following additional fees that may be required by this paper and during the entire pendency of this application to Account No. 12-0425.

☒ 37 C.F.R. 1.492(a)(1), (2), (3), and (4) (filing fees)

WARNING: *Because failure to pay the national fee within 30 months without extension (37 C.F.R. § 1.495(b)(2)) results in abandonment of the application, it would be best to always check the above box.*

☐ 37 C.F.R. 1.492(b), (c) and (d) (presentation of extra claims)

NOTE: *Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must*

only be paid or these claims cancelled by amendment prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 C.F.R. § 1.492(d)), it might be best not to authorize the PTO to charge additional claim fees, except possible when dealing with amendments after final action.

- ☒ 37 C.F.R. 1.17 (application processing fees)
☒ 37 C.F.R. 1.17(a)(1)-(5) (extension fees pursuant to § 1.136(a)).
☒ 37 C.F.R. 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. 1.311(b))

NOTE: Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance. 37 C.F.R. § 1.311(b).

NOTE: 37 C.F.R. 1.28(b) requires "Notification of any change in loss of entitlement to small entity status must be filed in the application . . . prior to paying, or at the time of paying . . . issue fee." From the wording of 37 C.F.R. § 1.28(b): (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.

- ☐ 37 C.F.R. § 1.492(e) and (f) (surcharge fees for filing the declaration and/or filing an English translation of an International Application later than 30 months after the priority date).



SIGNATURE OF PRACTITIONER

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1/pvt

02272US A METHOD OF GAINING METHYL ALCOHOL FROM RAW GAS AND APPARATUS
FOR PRACTICING THE METHOD

5

BACKGROUND OF THE INVENTION

The present invention relates to a method of
gaining methyl alcohol from a raw gas having contents of hy-
10 drogen sulfide and methane.

PRIOR ART

15 Natural gas occurs at many places on the earth
together with crude oil or also as pure natural gas. Pure
natural gas can be usually directly put to use, but in case
of mixtures, that is when natural gas occurs together with
crude oil, such is hardly possible. Generally, in case of
20 such mixtures the accompanying gas is flared off. By this a
certain amount of fossil fuel is wasted and at the same time
considerable emissions are produced.

The first alternative method of producing hydro-
carbon by a liquid process is based on the production of syn-
25 thetic gas from an incomplete combustion of coal (known as
Fischer/Tropsch-method 1925) to watergas which consists sub-
stantially of CO and H₂, which thereafter is synthesized via
a pressure catalysis to hydrocarbons. Further known, modern
methods such as Lurgi-Ruhrchemie-Kölbel-Reinpreussen, Kübel-
30 Engelhart and also further methods make use of additional oil
residues and produce aside from hydrocarbons also methanol.

All these methods have in common that they are
extremely expensive, complicated and necessitate an intensive

maintenance regarding the technical side of the plants, need a large amount of energy and are, therefore, not economical.

5

SUMMARY OF THE INVENTION

It is an object of the invention to provide a method of converting raw gas into a material which is simple and thus economically interesting.

10 The method according to the invention is characterized in that in a first cleaning step hydrogen-sulfide which is contained in the raw gas is separated by a microbial oxidation into water and sulfur, that the sulfur obtained is lead away and that thereafter in a second and third cleaning
15 step methane contained in the de-sulfurized raw gas is oxidized by a further microbial oxidation to methyl alcohol.

The apparatus for practicing the invention is characterized by a first sprinkled sprinkling body-solid bed-device with grown sulfide oxidizing bacteriae allocated to
20 the first cleaning step, in which sprinkling body-solid bed-device the raw gas is led in a counter flow to the washing liquid, which sprinkling body-solid bed-device includes a first washing water sump which communicates through a first sprinkling water conduit with a sprinkling nozzle arrangement
25 above the sprinkling body-solid bed-device, and which communicates through a draw-off conduit with a filter press with a following tank for the filtrate, from which tank a return circuit conduit runs back to a droplets removing device located at the outlet for the sulfurized gas, which draw-off
30 conduit has a connection to a polyelectrolyte source, which tank has a connection to a urea source, a connection to a phosphate acid source and a connection to a sodium liquor, whereby furthermore, a connection for a fresh air supply is

located in the space between the sprinkling body-solid bed and the washing water sump.

Advantageous embodiments are set forth in the dependent claims.

5

SHORT DESCRIPTION OF THE DRAWING

The subject of the invention will be described hereinbelow by example with reference to the drawing.

The single figure illustrates a circuit diagram of an apparatus for practicing the method according to the invention.

15

DESCRIPTION OF THE PREFERRED EMBODIMENT

Initially it shall be noted that the described method and correspondingly the described apparatus refer to the conversion of bio and raw gas containing hydrogen sulfide.

Firstly, the apparatus will be described. The apparatus includes three cleaning steps, namely a first cleaning step 1, a second cleaning step 17 and a third cleaning step 32. The first cleaning step includes a first sprinkling body-solid bed-device 2. It comprises a first sprinkling body-solid bed 6 and a first spray nozzle arrangement 5 for washing water is located above this sprinkling body-solid bed. A first washing water sump 3 is located under the first sprinkling body-solid bed. Washing water is led from the washing water sump 3 through a first sprinkling water line 4 to the first sprinkling nozzle arrangement. The raw gas to be treated enters at the inlet 59 into the space between the

first sprinkling body-solid bed 6 and the first washing water sump 3. Thus, it can be seen that the raw gas to be treated is led in a counter flow relative to the washing water. The first washing water sump 3 is connected through a draw-off conduit 7 to a chamber filter press 8. The reference numeral 13 designates a connection to a polyelectrolyte source. Thus, the washing liquid which flows through the draw-off line 7 is flocculated by polyelectrolyte so that it can be filtered off and dewatered in the following chamber filter press 8. A circulation conduit 8 extends from the tank 9 to a droplet remover 12 which is located at the area of the outlet 11 of the first cleaning step.

In this cleaning step 1 the de-sulfurizing of the hydrogen sulfides (H_2S) occurs in the inflowing raw gas. Sulfide oxidizing bacteriae have grown on the sprinkling body-solid bed 6. This solid bed 6 is continuously sprinkled by circulating water from the washing water sump 3. Through the connection 14 area for the nitrogen (N) supply, the connection 15 phosphoric acid for the phosphorous (P) supply and through the connection 16 an alkali, for instance sodium liquor or lime milk ($Ca, (OH)_2$) are added to the washing water sump 3. These additions are made in a dosed manner, thereby the reference numeral 60 designates a measuring point which measures the pH-value in order to execute a corresponding dosing of the added alkali.

Fresh air is fed into the space between the first sprinkling body-solid bed 6 and the first washing water sump 3 such as indicated by the arrow 22, so that a dosed addition of fresh air is arrived at. The dosing is, thereby, selected in such a manner that just the requirements of the sulfide oxidizing bacteriae are met. Because these bacteriae have a substantially higher oxidation potential than other bacteriae the selection proceeds automatically, i.e. it is ensured that

practically exclusively the sulfide oxidizing bacteriae remain in the sprinkling body-solid bed 6.

As a standard size for the dosing of the fed air or oxygen, respectively, the residual H_2S contents of the desulfurized gas coming out of the first sprinkling body-solid bed-device 1 can be taken.

The droplet remover 12 at the outlet 11 of the first cleaning step 1 keeps the escape of droplets within limits. The first oxidation step of the hydrogen sulfide is elementary sulfide corresponding to the relation $H_2S + O = H_2O + S$. Due to the deficiency of oxygen the elementary sulfur is not converted further but falls out fine dispersed in the washing liquid, is therefore, present in the first washing water sump 3. The washing water which contains the finely dispersed elementary sulfur flows from the first washing water sump 3 through the draw-off line 7 into the filter press 8. Such as already mentioned, the filtrate is collected in the tank 9 and is led back from there through the circulation conduit 10 back to the droplet remover 12.

Fresh water is added automatically in accordance with the level of the sprinkling body sump, i.e. washing water sump 36 and possible excess water from the filtrate tank 9 is drawn off.

The sulfur separated in the filter press is obtained in the form of a thick viscous cake which is identified generally by the reference numeral 61. It includes a minor bio mass content and can be utilized in the chemical industry, such as for the production of sulfuric acid or also as raw material in the cellulose, pulp industry. The sulfur is combusted, thereby, generally to SO_2 or SO_3 .

The desulfurized raw gas flows through the feeding conduit 19 to the second cleaning step 17. The reference numeral 20 denotes a connection for the feeding of fresh air.

In this second cleaning step 17 and the next following third cleaning step 32 an oxidation of methane occurs. Because now the sulfur has been separated out in the first cleaning step 1, it can not disturb the oxidation of the methane.

5 The second cleaning step 17 includes a second sprinkling body-solid bed-device 18. A second washing water sump is located under this sprinkling body-solid bed 25. A second sprinkling water line 24 extends from this second washing water sump 23 to the second sprinkling nozzle arrangement 38.

10 As can be seen, the raw gas enters the second cleaning step 17 at its upper section, i.e. above the second sprinkling body-solid bed 25, so that here the gas to be cleaned co-flows in the same direction as the sprinkling water. A mixed population of bacteriae and yeast has grown on the sprinkling body-solid bed 25, which mixed population performs predominantly the oxidation of methane.

15 The supply with nitrogen and phosphorous proceeds in the second washing water sump 23 through the connection 29 for a feeding of urea and a connection 30 for feeding phosphorus acid. The connection 31 designates the supply of an alkali, sodium liquor or also lime milk ($\text{Ca}(\text{OH})_2$) for the controlling of the pH-value. The measuring point for the measuring of the pH-value is identified by the reference numeral 21.

20 A washing water exit line 26 extends from the second washing water sump 23 to the third cleaning step 23. Again, a droplet remover 28 is located at the gas exit 27 of the second cleaning step.

30 A gas supply line 34 extends from this outlet 27 to the third cleaning step 32 which includes a third sprinkling body-solid bed-device 33 with a third sprinkling body-solid bed 40. The reference numeral 31 designates a connec-

tion for the supply of fresh air to the gas flowing towards the third cleaning step.

5 A third washing water sump 36 is located under the third sprinkling body-solid bed 40. A third sprinkling water line 37 extends from there to the third sprinkling nozzle arrangement 39.

Thus, as can be seen, the gas flows in the third cleaning step 32 in a counter flow relative to the washing water.

10 The reference numeral 41 designates at the third washing water sump 36 a connection to an alkali source, the connection 46 the connection to an urea source and the reference numeral 47 the connection to a phosphorous acid source. The reference numeral 62 designates the pH-measuring point.

15 The third washing water sump 36 communicates through a draw-off line 42 with a chamber-filter press 44. Again, a polyelectrolyte is added to this draw-off line 42 such as indicated by the connection 43, so that the washing water flowing to the filter press is flocculated. The filtrate coming out of the filter press 44 is collected in a
20 tank 45.

By the dosed adding of air (connection 35) and a control of the environmental conditions in the third cleaning step 32 a further oxidation of the methyl alcohol to formaldehyde and formic acid can be suppressed. In the circulating
25 water the methanol breaks down into an azeotrop. The contents on alcohol are not to be allowed to exceed 10 % because otherwise a toxic inhibiting of the microorganisms occurs. Thus, the dissolved, broken down methanol is continuously removed
30 from the circulating water.

Correspondingly, the third washing water sump 36 communicates through a draw-off line 42 with a filter press

44. The filtrate of the filter press 44 flows into the tank 45.

The solid matter which is arrived at in the filter press 44 and which consists substantially of microorganisms such as bacteriae, yeasts and mushrooms is separated out. The bio mass-produced is identified by the reference numeral 63.

This biomass is present in form of a dewatered excess sludge with a consistency of 20-40% dry matter. Because the biomass consists substantially of stabilized yeasts it can be used after a washing and a removing of the residual methanol as high-grade protein as a supplement in a fodder.

In the third cleaning step 32 CO₂, N₂ and (vaporuous) N₂O falls on, which gases and vapor, respectively, escape through the venting outlet 48.

The filtrate, namely water comprising 5-10 % methanol is collected in the tank 45 and led to a microfilter device 49. The separating and concentrating of the methanol proceeds now continuously in a distillation column 50. The heat needed for the evaporating in this distillation column 50 is supplied by an external heater 58. The liquid flowing away from the microfilter device 49 flows through the heat exchanger 54 and into the distillation column.

A liquid outlet line 53 extend from the distillation column 50 back to the heat exchanger 54 and a transfer line 55 extends from this heat exchanger 54 to a tank 56. A circulation line 57 extends from this tank 56 back to the droplet remover 28 of the second treatment step. Thus, the washing water is led in a circuit between the second treatment step 17 and the third treatment step 32.

The distillation vapor is led from the distillation column 50 to a cooled condenser 51 and is correspondingly condensated. The methanol distillate is finally col-

lected in the tank 52. This single-distillate features a methanol content of 30-40 %. Higher contents can obviously be produced by a rectification.

Now, methanol (methyl alcohol) has been gained
5 from the original raw gas. Methanol is a basic raw material for the chemical industry. The production amounts for instance in Germany to about 700'000 t/year. Methanol (CH_3OH) is a watery, colorless, burnable liquid, can be ignited easily and burns with a bluish flame to CO_2 and H_2O . Possible
10 applications for methanol are an alternative Otto-fuel, a admixing component for gasoline, catalytic transformation to gasoline, synthetic raw material for the chemical industry. Methanol can be, furthermore, easily split up to a "reformer" in CO_2 and H_2 ; by means of this a direct supply of fuel cells
15 is possible.

Because the method operates with the employment of microorganisms and the sulfur which would disturb the methane oxidation is separated out in the first step, this method displays a high economy and is simultaneously not disturbing the environment. The expenditure of energy and chemicals is low and the apparatus can be set up in a modular design at any location where the gas occurs and can be automated to a large extent.
20

25

CLAIMS

1. A method of gaining methyl alcohol from a raw gas which contains hydrogen sulfide and methane, characterized in that in a first claiming step hydrogen sulfide which is present in the raw gas is separated by a microbial oxidation into water and sulfur and the sulfur obtained is led away, and that thereafter in a second and third cleaning step methane which is present in the desulfurized raw gas is oxidized by a further microbial oxidation to methyl alcohol.

2. The method of claim 1, characterized in that the hydrogen sulfide is oxidized in a first sprinkling body-solid bed onto which water conducted in a circuit is sprinkled by sulfide oxidizing bacteriae, and in that for the supplying with nitrogen and phosphorous urea and phosphoric acid and for a maintaining of the pH-value an alkali are fed dosed to the circulating water.

3. The method of claim 1, characterized in that oxygen is fed in a limited amount by a dosed adding of fresh air to the raw gas, which amount covers not more than the requirement of the sulfide oxidizing bacteriae, whereby the H₂S-content of the raw gas exiting the first sprinkling body-solid bed is measured and taken as the standard value for the adding of the fresh air.

4. The method of claim 2, characterized in that washing liquid is discontinuously drawn off the circulating water, is flocculated by polyelectrolyte and filtered out and denatured in a filter press, and the filtrate is led back into the circulating water.

5. The method of claim 1, characterized in that the microbial oxidation of the methane in the second and

third cleaning step proceeds in both in a sprinkling body-solid bed with a mixed population of bacteriae and yeasts by water led in a circuit, whereby for a supply with nitrogen and phosphorous urea and phosphoric acid and for a maintaining of the pH-value an alkali are fed to the circulating water of both cleaning steps.

6. The method of claim 5, characterized in that by a dosed adding of fresh air oxygen is added in a limited amount to the desulfurized raw gas ahead of the respective entries into a sprinkling body-solid bed, which amount does not exceed the requirement of the mixed population of methane oxidizing bacteria and yeasts in order to suppress a further oxidation of the gained methyl alcohol to formaldehyde by formic acid, whereby the methane content of the pure gas after the exit out of the sprinkling body-solid bed of the third cleaning step is measured and taken as standard value for the adding of air.

7. The method of claim 6, characterized in that depending from the measured content of methane of the pure gas in both steps washing liquid is alternately drawn off the circulating water, is flocculated by polyelectrolyte and is freed in a filter press and by micro filtration from solid matter.

8. The method of claim 7, characterized in that in order to separate and to concentrate the methanol the methanol containing water is heated after the micro filtration, distilled in a distillation column and condensated in a condenser, and the distillate is led to a product container.

9. Apparatus for practicing the method of claim 1, characterized by a first sprinkled sprinkling body-solid bed-device with grown sulfide oxidizing bacteriae allocated to the first cleaning step, in which sprinkling body-solid bed-device the raw gas is led in a counter current relative

to the washing liquid, which sprinkling body-solid bed-device includes a first washing water sump which communicates through a first sprinkling water line with a first sprinkling nozzle arrangement over the sprinkling body-solid bed of the first sprinkling body-solid bed-device, and which communicates through a draw-off line with a filter press with a following tank for the filtrate, from which tank a circulation line extends to a droplets remover located at the outlet for the desulfurized raw gas, which draw-off line has a connection to a polyelectrolyte source, which first washing water sump has a connection to an urea source, a connection to a phosphoric acid source and a connection to an alkali source, whereby furthermore a connection for the feeding of fresh air is located at the space between the sprinkling body-solid bed and the washing water sump.

10. Apparatus according to claim 9, characterized by a second sprinkled sprinkling body-solid bed-device with a second sprinkling body-solid bed with a grown mixed population of bacteriae and yeasts for performing the methane oxidation allocated to the second cleaning step, which second sprinkling body-solid bed-device communicates through a feeding line with a connection for feeding of fresh air with the outlet of the first sprinkling body-solid bed-device, in which second sprinkling body-solid bed-device the desulfurized raw gas is led in a co-flow relative to the washing liquid, which second sprinkling body-solid bed-device has a second washing water sump, which communicates through a second sprinkling water line with a second sprinkling water arrangement over the second sprinkling body-solid bed, to which second washing water sump a washing water outlet line is connected, whereby a droplet remover is located between the second sprinkling body-solid bed and the second washing water sump at the outlet for the treated gas, and whereby the sec-

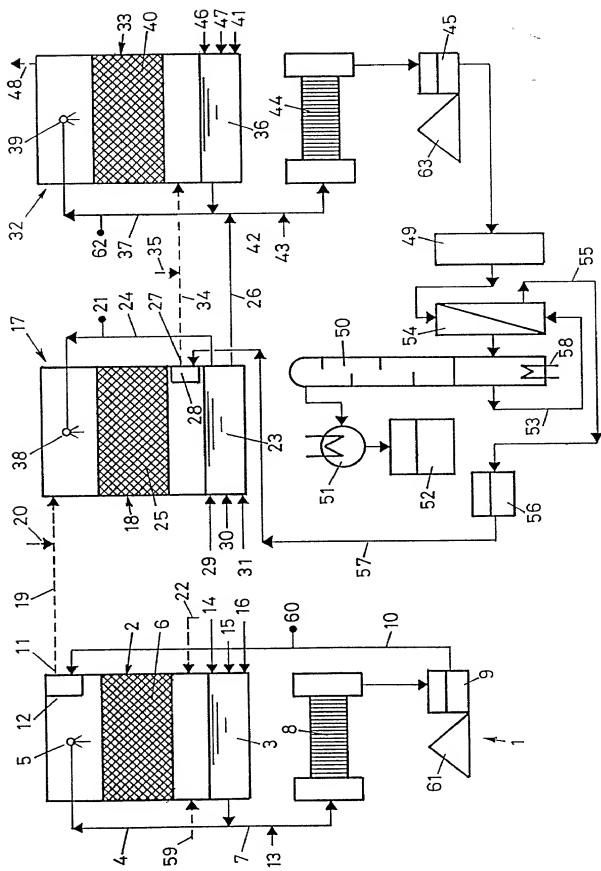
ond washing water sump has a connection to an urea source, a connection to a phosphorous acid and a connection to an alkali source.

11. Apparatus according to claim 10, characterized by a third sprinkled sprinkling body-solid bed-device with a third sprinkling body-solid bed with a grown mixed population of bacteria and yeasts for a final oxidation of methane allocated to the third cleaning step, which third sprinkling body-solid bed-device communicates through a feeding line with a connection for the supply of fresh air with the outlet for the treated gas of the second sprinkling body-solid bed-device, in which third sprinkling body-solid bed-device the gas to be treated is led in a counter-current relative to the washing liquid, which third sprinkling body-solid bed-device has a third washing water sump which communicates through a third sprinkling water line with a sprinkling nozzle arrangement over the third sprinkling body-solid bed, which third sprinkling water line has a connection to an alkali source, from which sprinkling water line a draw-off line is branched off, to which draw-off line the washing water exit line of the second washing water sump of the second sprinkling body-solid bed-device is led, has a connection extending to an electrolyte source and to a filter press with a following tank for the filtrate, which third washing water sump has a connection to an urea source, a connection to a source of phosphorous acid and a connection to an alkali source, and which third sprinkling body-solid bed-device has a ventilating outlet above the sprinkling body-solid bed.

12. Apparatus according to claim 11, characterized by a microfilter device following regarding the direction of flow of the washing water the tank of the third sprinkling body-solid bed-device, and which is followed by a heat exchanger and a heated distillation column, which is

followed by a condenser and a tank for the distillate which contains the distillate.

13. Apparatus according to claim 12, characterized in that a liquid outlet line extends from the distillation column to the heat exchanger which is connected, furthermore, through a transfer line to a tank, from which tank a circulation line extends to the droplets remover of the second sprinkling body-solid bed-device.



Optional Customer No. Bar Code



00140

PATENT TRADEMARK OFFICE

COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL,
CONTINUATION, OR C-I-P)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

(check one applicable item below)

- ☐ original.
☐ design.

NOTE: With the exception of a supplemental oath or declaration submitted in a reissue, a supplemental oath or declaration is not treated as an amendment under 37 CFR 1.312 (Amendments after allowance). M.P.E.P. Section 714.16, 7th Ed.

- ☐ supplemental.

NOTE: If the declaration is for an International Application being filed as a divisional, continuation or continuation-in-part application, do not check next item; check appropriate one of last three items.

- ☒ national stage of PCT.

NOTE: If one of the following 3 items apply, then complete and also attach ADDED PAGES FOR DIVISIONAL, CONTINUATION OR C-I-P.

NOTE: See 37 C.F.R. Section 1.63(d) (continued prosecution application) for use of a prior nonprovisional application declaration in the continuation or divisional application being filed on behalf of the same or fewer of the inventors named in the prior application.

- ☐ divisional.
☐ continuation.

NOTE: Where an application discloses and claims subject matter not disclosed in the prior application, or a continuation or divisional application names an inventor not named in the prior application, a continuation-in-part application must be filed under 37 C.F.R. Section 1.53(b) (application filing requirements-nonprovisional application).

- ☐ continuation-in-part (C-I-P).

INVENTORSHIP IDENTIFICATION

WARNING: *If the inventors are each not the inventors of all the claims, an explanation of the facts, including the ownership of all the claims at the time the last claimed invention was made, should be submitted.*

My residence, post office address and citizenship are as stated below, next to my name. I believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter that is claimed, and for which a patent is sought on the invention entitled:

TITLE OF INVENTION

METHOD OF OBTAINING METHYL ALCOHOL FROM RAW GAS AND INSTALLATION
FOR CARRYING OUT SAID METHOD

SPECIFICATION IDENTIFICATION

The specification of which:

(complete (a), (b), or (c))

(a) ☐ is attached hereto.

NOTE: *"The following combinations of information supplied in an oath or declaration filed on the application filing date with a specification are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F.R. Section 1.63:*

"(1) name of inventor(s), and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration on filing;

"(2) name of inventor(s), and attorney docket number which was on the specification as filed; or

"(3) name of inventor(s), and title which was on the specification as filed."

Notice of July 13, 1995 (1177 O.G. 60).

(b) ☒ was filed on September 4, 2001, ☐ as Application No. _____
☐ and was amended on _____ (if applicable).

NOTE: *Amendments filed after the original papers are deposited with the PTO that contain new matter are not accorded a filing date by being referred to in the declaration. Accordingly, the amendments involved are those filed with the application papers or, in the case of a supplemental declaration, are those amendments claiming matter not encompassed in the original statement of invention or claims. See 37 C.F.R. Section 1.67.*

NOTE: *"The following combinations of information supplied in an oath or declaration filed after the filing date are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 C.F.R. Section 1.63:*

(A) application number (consisting of the series code and the serial number, e.g., 08/123,456);

(B) serial number and filing date;

(C) attorney docket number which was on the specification as filed;

(D) title which was on the specification as filed and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration; or

(E) title which was on the specification as filed and accompanied by a cover letter accurately identifying the application for which it was intended by either the application number (consisting of the series code and the serial number, e.g., 08/123,456), or serial number and filing date. Absent any statement(s) to the contrary, it will be presumed that the application filed in the PTO is the application which the inventor(s) executed by signing the oath or declaration.

M.P.E.P. Section 601.01(a), 7th ed.

- (c) ☒ [X] was described and claimed in PCT International Application No. IB00/00290 filed on March 17, 2000 and as amended under PCT Article 19 on _____ (if any).

SUPPLEMENTAL DECLARATION (37 C.F.R. Section 1.67(b))

(complete the following where a supplemental declaration is being submitted)

☐ [] I hereby declare that the subject matter of the

☐ [] attached amendment

☐ [] amendment filed on _____.

was part of my/our invention and was invented before the filing date of the original application, above identified, for such invention.

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, Section 1.56,

(also check the following items, if desired)

☐ [] and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent, and

☐ [] in compliance with this duty, there is attached an information disclosure statement, in accordance with 37 C.F.R. Section 1.98.

PRIORITY CLAIM (35 U.S.C. Section 119(a)-(d))

NOTE: "The claim to priority need be in no special form and may be made by the attorney or agent if the foreign application is referred to in the oath or declaration as required by Section 1.63. The claim for priority and the certified copy of the foreign application specified in 35 U.S.C. Section 119(b) must be filed in the case of an interference (Section 1.630), when necessary to overcome the date of a reference relied upon by the examiner, when specifically required by the examiner, and in all other situations, before the patent is granted. If the claim for priority or the certified copy of the foreign application is filed after the date the issue fee is paid, it must be accompanied by a petition requesting entry and by the fee set forth in Section 1.17(i). If the certified copy is not in the English language, a translation need not be filed except in the case of interference; or when necessary to overcome the date of a reference relied upon by the examiner; or when specifically required by the examiner, in which event an English language translation must be filed together with a statement that the translation of the certified copy is accurate." 37 C.F.R. Section 1.55(a).

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(complete (d) or (e))

- (d) ☐ no such applications have been filed.
(e) ☒ such applications have been filed as follows.

NOTE: Where item (c) is entered above and the International Application which designated the U.S. itself claimed priority check item (e), enter the details below and make the priority claim.

**PRIOR FOREIGN/PCT APPLICATION(S) FILED WITHIN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS APPLICATION
AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. SECTION 119(a)-(d)**

COUNTRY (OR INDICATE IF PCT)	APPLICATION NUMBER	DATE OF FILING DAY, MONTH, YEAR	PRIORITY CLAIMED UNDER 35 USC 119
CH	507/99	18 March 1999	<input checked="" type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO
			<input type="checkbox"/> YES <input type="checkbox"/> NO

**CLAIM FOR BENEFIT OF PRIOR U.S. PROVISIONAL APPLICATION(S)
(35 U.S.C. Section 119(e))**

I hereby claim the benefit under Title 35, United States Code, Section 119(e) of any United States provisional application(s) listed below:

PROVISIONAL APPLICATION NUMBER

FILING DATE

**CLAIM FOR BENEFIT OF EARLIER U.S./PCT APPLICATION(S)
UNDER 35 U.S.C. SECTION 120**

- ☐ The claim for the benefit of any such applications are set forth in the attached
ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY
FOR DIVISIONAL, CONTINUATION OR CONTINUATION-IN-PART (C-I-P)
APPLICATION.

ALL FOREIGN APPLICATION(S), IF ANY, FILED MORE THAN 12 MONTHS
(6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

NOTE: If the application filed more than 12 months from the filing date of this application is a PCT filing forming the basis for this application entering the United States as (1) the national stage, or (2) a continuation, divisional, or continuation-in-part, then also complete ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR C-I-P APPLICATION for benefit of the prior U.S. or PCT application(s) under 35 U.S.C. Section 120.

POWER OF ATTORNEY

I hereby appoint the following practitioner(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

(list name and registration number)

JOSEPH H. HANDELMAN, 26179

JULIAN H. COHEN, 20302

JOHN RICHARDS, 31053

WILLIAM R. EVANS 25858

RICHARD J. STREIT, 25765

JANET I. CORD, 33778

PETER D. GALLOWAY, 27885

CLIFFORD J. MASS, 30086

IAIN C. BAILLIE, 24090

CYNTHIA R. MILLER, 34678

RICHARD P. BERG, 28145

(Check the following item, if applicable)

- ☐ I hereby appoint the practitioner(s) associated with the Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.
- ☐ Attached, as part of this declaration and power of attorney, is the authorization of the above-named practitioner(s) to accept and follow instructions from my representative(s).

NOTE: "Special care should be taken in continuation or divisional applications to ensure that any change of correspondence address in a prior application is reflected in the continuation or divisional application. For example, where a copy of the oath or declaration from the prior application is submitted for a continuation or divisional application filed under 37 CFR 1.53(b) and the copy of the oath or declaration from the prior application designates an old correspondence address, the Office may not recognize, in the continuation or divisional application, the change of correspondence address made during the prosecution of the prior application. Applicant is required to identify the change of correspondence address in the continuation or divisional application to ensure that communications from the Office are mailed to the current correspondence address. 37 CFR 1.63(d)(4)." Section 601.03, M.P.E.P., 7th Ed

SEND CORRESPONDENCE TO

Ladas & Parry
26 West 61st Street
New York, N.Y. 10023

DIRECT TELEPHONE CALLS TO:

(Name and telephone number)

William R. Evans
(212) 708-1930

(complete the following if applicable)

Since this filing is a [] continuation [] divisional there is attached hereto a Change of Correspondence Address so that there will be no question as to where the PTO should direct all correspondence.

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURE(S)

NOTE: Carefully indicate the family (or last) name, as it should appear on the filing receipt and all other document.

NOTE: Each inventor must be identified by full name, including the family name, and at least one given name without abbreviation together with any other given name or initial, and by his/her residence, post office address and country of citizenship. 37 C.F.R. Section 1.63(a)(3).

NOTE: Inventors may execute separate declarations/oaths provided each declaration/oath sets forth all the inventors. Section 1.63(a)(3) requires that a declaration/oath, inter alia, identify each inventor and prohibits the execution of separate declarations/oaths which each sets forth only the name of the executing inventor. 62 Fed. Reg. 53,131, 53,142, October 10, 1997.

Full name of sole or first inventor

1-00
EMIL _____ A.J. _____ WIESER-LINHART _____
(Given Name) (Middle Initial or Name) Family (Or Last Name)
Inventor's signature (X) Emil A.J. Wieser-Linhart 28th Sept. 01
Date (X) _____ Country of Citizenship AUSTRIA
Residence DOSENWEG 49, A-5020 SALZBURG, AUSTRIA ATX
Post Office Address SAME AS ABOVE

Full name of second joint inventor, if any

(Given Name) _____ (Middle Initial or Name) _____ Family (Or Last Name) _____
Inventor's signature _____
Date _____ Country of Citizenship _____
Residence _____
Post Office Address _____

Full name of third joint inventor, if any

(Given Name) _____ (Middle Initial or Name) _____ Family (Or Last Name) _____
Inventor's signature _____
Date _____ Country of Citizenship _____
Residence _____
Post Office Address _____

(check proper box(es) for any of the following added page(s)
that form a part of this declaration)

☐ **Signature** for fourth and subsequent joint inventors. *Number of pages added* _____

* * *

☐ **Signature** by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. *Number of pages added* _____

* * *

☐ **Signature** for inventor who refuses to sign or cannot be reached by person authorized under 37 C.F.R. Section 1.47. *Number of pages added* _____

* * *

☐ Added page for **signature** by one joint inventor on behalf of deceased inventor(s) where legal representative cannot be appointed in time. (37 C.F.R. Section 1.47)

* * *

☐ Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (C-I-P) application.
[] Number of pages added _____

* * *

☐ Authorization of practitioner(s) to accept and follow instructions from representative.

(If no further pages form a part of this Declaration,
then end this Declaration with this page and check the following item)

☒ This declaration ends with this page.